O 70: Poster: Spins on Surfaces & Surface Magnetism

Time: Wednesday 18:00–20:00 Location: Poster C

O 70.1 Wed 18:00 Poster C

Weakly coupled S=1/2 molecules on a superconductor — •Vivek Krishnakumar Rajathilakam[1], Nicolaj Betz[1], Lukas Arnhold[1], Matteo Briganti[2], Lorenzo Poggini[4], Jaisa Fernandez Soarez[5], Luana C. De Camargo[5], Giulia Serrano[3], Matteo Mannini[2], Federico Totti[2], Roberta Sessoli[2], Susanne Baumann[1], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Department of Chemistry Ugo Schiff, University of Florence, Italy — [3]Department of Industrial Engineering, University of Florence, Italy — [4]Institute of Chemistry of Organometallic Compounds - CNR, Florence, Italy — [5]Department of Chemistry, Federal University of Paraná, Brazil

Sandwich complexes hosting a transition metal ion can show magnetic properties of great interest for quantum information processing [1]. Here we study an S=1/2 organometallic sandwich CpTi(cot), with very long coherence times in frozen solution [2]. We use a Scanning Tunneling Microscope (STM) at 50 mK to measure the properties of individual CpTi(cot) molecules adsorbed on a superconducting vanadium surface V(100) with oxygen reconstruction. It shows no Yu-Shiba-Rusinov states, but prominent S=1/2 spin excitations can be traced up to 15 T. They show spin-spin interactions for molecules with separation up to 1.4 nm. These results show a promising step towards using molecules on superconductors for molecular qubit applications. [1] AM. Ariciu, et al. Nat Commun 10, 3330 (2019) [2] L.C. de Camargo, et al. Angew. Chem., Int. Ed., 60, 2588 (2021)

O 70.2 Wed 18:00 Poster C

Controlling Yu-Shiba-Rusinov States with a Molecular Switch — •Arnab Banerjee, Niklas Ide, Richard Berndt, and Alexander Weismann — Instituts für Experimentelle und Angewandte Physik, CAU Kiel

Tin phthalocyanine (SnPc) is a non-planar diamagnetic molecule. On metal surfaces, it is geometrically bistable and transitions may be induced by electron injection. We studied SnPc on superconducting Pb(100) using low-temperature scanning tunneling spectroscopy. Yu-Shiba-Rusinov (YSR) states are observed upon assembly of the molecules into supramolecular arrays indicating the presence of a molecular spin. Moreover, the effect of geometrical switching on the YSR state is demonstrated.

O 70.3 Wed 18:00 Poster C

Quantum simulator to emulate lower dimensional physics and chemistry — Emil Sierda, Xiaochun Huang, Danis Badrtdinov, Brian Kiraly, Elze Knol, •Anna M. H. Krieg, Gerrit C. Groenenboom, Mikhail I. Katsnelson, Malte Rösner, Daniel Wegner, and Alexander A. Khajetoorians — Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands

Designing materials with tailored physical and chemical properties requires a quantitative understanding of interacting quantum systems. In order to provide predictability, a promising route is to create bottom-up platforms, where the electronic properties of individual and interacting atoms can be emulated in a tunable manner. Here, we present a quantum simulator based on patterned Cs ions embedded in a 2DEG on the surface of semiconducting InSb(110). We use this platform to emulate the structure and orbital landscape of planar organic molecules. Using STM/STS and ab initio calculations, we show that an artificial atom can be derived from localized states of patterned Cs ions. The resultant potential can be used to couple artificial atoms, leading to bonding and anti-bonding states as well as to different orbital symmetries. Based on these artificial orbitals, we emulate molecular orbitals of known organic molecules based on various atomic structures. In a different limit where Cs atoms are much closer, this quantum simulator can also be used to probe the effect of e-e interactions. Our experimental data suggest that dense structures exhibit many-body effects which can be extended to complex quantum states based on arbitrary lattices.

O 70.4 Wed 18:00 Poster C

Non-contact layer stabilization of azafullerene radicals: route towards high-spin-density surfaces — Yuri Tanuma[1,2], Gregor Kladnik[3,4], Luca Schio[4], •Marion van Midden Mavrič[1], Bastien Anézo[5], Erik Zupanič[1], Gregor Bavdek[4,6], Ruben Canton-Vitoria[7], Luca Floreano[4], Nikos Tagmatarchis[7], Hermann A. Wegner[8,9], Alberto Morgante[4,10], Chris Ewels[5], Dean Cvetko[1,3,4], and Denis Arčon[1,3] — [1]Jožef Stefan Institute, Ljubljana, Slovenia — [2]CAREM, Hokkaido University, Sapporo, Japan — [3]Faculty of Mathematics and Physics, University of Ljubljana, Slovenia — [4]CNR-IOM. Trieste, Italy — [5]IMN, Nantes, France — [6]Faculty of Education, University of Ljubljana, Slovenia — [7]Theoretical and Physical Chemistry Institute, Athens, Greece — [8]Institute of Organic Chemistry, Giessen, Germany — [9]Center for Materials research, Giessen, Germany — [10]Physics department, University of Trieste, Italy

Organic radicals offer a promising qubit platform. Despite them usually being unstable, as their unpaired electrons are highly reactive, they can be stabilized via weak interactions with the surface.

We study $C_{59}N^{\bullet}$ deposited on Au(111) in UHV at molecular coverage up to 2ML. Using NEXAFS and STM we show that single azafullerenes are indeed deposited on the surface. Together with DFT calculations, NEXAFS/XPS allows us to identify the spectroscopic fingerprint of the molecular radical state. The binding of $C_{59}N^{\bullet}$ changes as a function of coverage. We identify a novel intermediate high-spin-density state between 1ML and 2ML.

O 70.5 Wed 18:00 Poster C

Manipulating Quantum States: Indirect Readout of ^{163}DyPc$_2$ Single-Molecule Magnets via Spin Cascade on Au(111) using Scanning Tunneling Microscopy — •Simon Gerber[1], Hongyan Chen[1], Philip Schmid[1], Vera Schmeiser[1], Svetlana Klyatskaya[1], Eufemio Moreno-Pineda[2], Mario Ruben[1,3], and Wulf Wulfhekel[1] — [1]Karlsruher Institute of Technology — [2]Universidad de Panamá — [3]Centre Européen de Sciences Quantiques, Straßbourg

We present an innovative method to indirectly read the nuclear spin of ^{163}DyPc$_2$ (I=5/2) single-molecule magnets on Au(111) using milli-Kelvin scanning tunneling microscopy. The Kondo effect, initiated by an unpaired electron in the π-radical on the exposed Pc ligand, induces a spin cascade[1,2], revealing discrete signal jumps on the order of minutes. These jumps, which may be correlated with nuclear spin flips, suggest possibilities for manipulation with an RF source. First results of RF-induced transitions are presented. This study focuses on the interplay between local 4f electrons, unpaired electrons and nuclear spins in rare earth-based systems. We demonstrate that reading the nuclear spin of ^{163}DyPc$_2$ using spin-polarised scanning tunneling microscopy (Sp-STM) does not need a magnetic field. This research makes a significant contribution to the understanding and manipulation of quantum states in single molecule magnets.

[1] R. Vincent et al,. Nature **488**, 357 (2012); S. Thiele et al., Science **344**, 1135 (2014) [2] T. Frauhammer, H. Chen et al., Phys. Rev. Lett. **127**, 123201 (2021)

O 70.6 Wed 18:00 Poster C

Mn on 2H-NbSe2 - From Single Magnetic Atoms to Spin Chains — •Margarete Huisinga[1], Lisa M. Rütten[1], Kai Rossnagel[2,3], and Katharina J. Franke[1] — [1]Fachbereich Physik, Freie Universität Berlin, Germany — [2]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany — [3]Ruprecht Haensel Laboratory, Deutsches Elektronen- Synchrotron DESY, Hamburg, Germany

On superconductors the interaction of unpaired electron spins of magnetic atoms with the Cooper pairs gives rise to so called Yu-Shiba-Rusinov (YSR) states. These states can be probed on the atomic scale using a scanning tunneling microscope (STM). YSR states are influenced by the environment of the atom on the surface such as its position on the lattice or a charge density wave (CDW).

Here, we investigate Mn atoms on 2H-NbSe2. Using the tip of a STM we build chains of Mn atoms. Interestingly, densely packed Mn chains exhibit inelastic excitations outside the superconducting gap along with YSR states. These inelastic excitations depend on the parity of the number of atoms in the chain, and vary in intensity and energy along the chain. We investigate the evolution of the inelastic excitations and YSR states with increasing chain length.

O 70.7 Wed 18:00 Poster C

Controlling hybridization in Yu-Shiba-Rusinov dimers through a charge-density wave — •Lisa M. Rütten[1], Eva Liebhaber[1], Kai Rossnagel[2,3], and Katharina J. Franke[1] — [1]Fachbereich Physik, Freie Universität Berlin, Germany — [2]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany — [3]Ruprecht Haensel Laboratory, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany

Unpaired adatom spins on superconductors interact with the Cooper pairs of the substrate and induce Yu-Shiba-Rusinov (YSR) states inside the superconducting gap. These states can be probed by scanning tunneling spectroscopy at the single-atom scale. In quasi-two-dimensional materials, the YSR wave functions extend over several nanometers. If the YSR wave functions of two impurities overlap they can hybridize which is observed as a splitting of YSR resonances in tunneling spectra. Additionally, the YSR states are influenced by the presence of a charge-density wave (CDW). Fe atoms on $2H$-NbSe$_2$ have been shown to exhibit long-range YSR wave functions, where the energies, at which the YSR resonances are observed, depend strongly on the atoms position with respect to the CDW.

Here, we use lateral manipulation to arrange Fe atoms on $2H$-NbSe$_2$. While an isolated atom hardly influences the CDW, a set of atoms may impact its incommensurate pattern. We build a dimer of Fe atoms and then rearrange additional atoms around it to modify the CDW. We show that the hybridization strength can be switched as the underlying CDW is changed.

O 70.8 Wed 18:00 Poster C

Chiral hybridization of Yu-Shiba-Rusinov states in Fe clusters on Pb(111) — •Bharti Mahendru[1], Léa Meynier[1], Lisa M. Rütten[1], Harald Schmid[1], Felix von Oppen[1,2], and Katharina J. Franke[1] — [1]Fachbereich Physik, Freie Universität Berlin, Berlin, Germany — [2]Dahlem Center for Complex Quantum Systems and Fachbereich Physik, Freie Universität Berlin, Berlin, Germany

Unpaired electron spins exchange coupled to a superconductor give rise to bound states inside the superconducting energy gap, so-called Yu-Shiba-Rusinov (YSR) states. Scanning tunneling microscopy and spectroscopy (STM/STS) are powerful tools to probe the excited state around the Fermi level of such systems at the atomic scale. Previously, it has been shown that the crystal field splits the d-levels of Mn atoms on Pb surfaces, which lead to distinct YSR states inheriting the symmetry of the spin-carrying orbital. Here, we investigate self-assembled clusters of Fe on Pb(111) using STM/STS. Single Fe atoms, although having four unpaired electrons in the d-levels in gas phase, do not show YSR states on Pb(111) surface. In contrast, we observe different clusters, all showing in-gap states. These YSR states exhibit different symmetries that are related to the structure of the cluster on the Pb(111) surface. Interestingly, one type of the cluster exhibits spatially chiral YSR wavefunctions in differential conductance (dI/dV) maps. Using a simple model, we find that the origin of this chirality results from the adsorption of Fe atoms in the three-fold symmetric hollow sites of the substrate and the extent of the YSR wavefunctions.

O 70.9 Wed 18:00 Poster C

Self-Assembly of Magnetic Atom-Molecule Complexes on a Surface — Wantong Huang[1], Paul Greule[1], Máté Stark[1], Joris Van Slageren[2], Benedetta Casu[3], Christoph Sürgers[1], Wolfgang Wernsdorfer[1], Giorgio Sangiovanni[4], Christoph Wolf[5], and •Philip Willke[1] — [1]Physikalisches Institut, Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Institute of Physical Chemistry and Center for Integrated Quantum Science and Technology IQST, University of Stuttgart, Pfaffenwaldring 55, 70569 Stuttgart, Germany — [3]Institute of Physical and Theoretical Chemistry, University of Tübingen, Auf der Morgenstelle 18, 72076 Tübingen, Germany. — [4]Institut für Theoretische Physik und Astrophysik, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [5]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, Republic of Korea.

Both single atomic as well as molecular spin centers are prime candidates for atomic-scale quantum information processing and often individually addressed and manipulated using STM. Here, we demonstrate a reliable self-assembly of magnetic complexes consisting of individual iron (Fe) atoms and molecules on a silver substrate. We employ two types of molecules, bis(dibenzoylmethane) copper(II) [Cu(dbm)2] and iron phthalocyanine (FePc), and show that Fe atoms always preferentially attach underneath the benzene ring ligand of the molecules. In both cases, a molecule can host up to two Fe atoms. In addition, we observe a change in the magnetic properties of the attached Fe atoms in spectroscopy, revealing a distinct Kondo signature at the Fe site.

O 70.10 Wed 18:00 Poster C

Exploring the magnetic field dependence of emulated molecular orbitals on Cs/InSb(110) — •Hermann Osterhage, Niels P. E. van Mullekom, Emil Sierda, Kira Junghans, Julian H. Strik, Daniel Wegner, and Alexander A. Khajetoorians — Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands

Quantum simulators can be used to emulate custom-made Hamiltonians that are mathematically hard to solve in a physical system. In a recent realization of a quantum simulator, molecular orbitals were emulated by positioning individual Cs atoms on an InSb(110) surface [1]. It was previously shown that this material system also exhibits Landau quantization in the presence of an out-of-plane magnetic field [2]. On this platform, artificial atoms can be realized as localized electronic states created from ring structures of Cs atoms, assembled using scanning tunneling microscopy. Coupling these artificial atoms in different geometries results in artificial molecular structures that emulate planar organic molecules with different symmetries [1]. The resulting structures show electronic states of multi-orbital, as well as hybrid orbital character. Here, we explore the role of magnetic field on the emulated molecular states.

[1] E. Sierda *et al.*, Science 380, 1048 (2023).
[2] K. Hashimoto *et al.*, Phys. Rev. Lett. 101, 256802 (2008).

O 70.11 Wed 18:00 Poster C

Simulation of the nuclear spin state switching driven by ESR in STM — •Hester Vennema[1], Rik Broekhoven[1], Evert Stolte[1], Esther Teng[1], Jinwon Lee[1], Philip Willke[2], and Sander Otte[1] — [1]Delft University of Technology, Delft, The Netherlands — [2]Karlsruhe Institute of Technology, Karlsruhe, Germany

The spin of a single nucleus is a prime candidate for quantum information applications due to its weak coupling to the environment and subsequently longer coherence times. Using the high energy resolution of in combination with the high spatial resolution of STM, it is possible to measure the hyperfine interaction between the nucleus and an electron for single atoms on a surface. A recent effort has been proven successful to resolve the coherent dynamics of a hyperfine driven interaction between nucleus and electron spin.

In our latest experiment, we have observed nuclear spin switching in real time of a 49Ti isotope (I = 7/2) on MgO/Ag(100). The electron spin is continuously driven by a constant RF voltage and interacts with the nucleus by hyperfine interaction. In this work, we demonstrate a model to gain insight in the seen dynamics. Particularly, we are interested in the effect on the nuclear spin dwell time when driving the electron spin slightly off resonance. Combining open system dynamics for two coupled spins, and an intuitive simulation of the experiment, we gain insight in the interaction of the electron and the nucleus. This could lead to understanding dominant relaxation processes and improving conditions to increase the lifetime of the nuclear spin.

O 70.12 Wed 18:00 Poster C

Magnetic adatoms on ultrathin films of the hexagonal insulator ZnO — •Henrik Lichtl[1], Lukas Arnhold[1], Leon Rullkoetter[1], Nicolaj Betz[1], Christoph Wolf[2], Susanne Baumann[1], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Center for Quantum Nanoscience, Institute for Basic Science, Seoul, South Korea

Different thin insulating layers can be used to tailor the electronic properties of adsorbed species. We use a double layer of zinc oxide (ZnO), a hexagonal wide bandgap semiconductor[1], to mitigate electron scattering between the Ag (111) substrate and Co atoms deposited on the ZnO surface. Ultrathin zinc oxide (ZnO) forms a honeycomb lattice and a Moiré superlattice on Ag(111). Scanning tunneling microscopy (STM) and density-functional theory (DFT) show that the Co atoms adsorb in the hollow sites of the ZnO hexagons. The atoms can be manipulated laterally with the STM tip. We find different Co entities that range from individual atoms to small nanostructures that feature distinct shape- and magnetic field-dependent spin excitations. This study lays the foundation for further exploration and manipulation of spins on ZnO towards design of frustrated atomic-scale magnetic systems.

[1] A. Shiotari, et al., J. Phys. Chem. C 2014, 118, 47, 27428-27435

O 70.13 Wed 18:00 Poster C

Coherent control of artificial atom-molecule hybrids — •Máté Stark[1], Wantong Huang[1], Paul Greule[1], Daria Sostina[2], Constantin Weeber[1], Jose Gálvez[3], Christoph Sürgers[1], Christoph Wolf[3], Wolfgang Wernsdorfer[1], and Philip Willke[1] — [1]Physikalisches Institut (PHI), Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Institute of Quantum Materials and Technologies (IQMT), Karlsruhe Institute of Technology, Karlsruhe, Germany — [3]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, Republic of Korea

By harnessing the atomic precision of Scanning Tunneling Microscopy (STM) in tandem with Electron Spin Resonance (ESR), we show in this work that artificially built atom-molecule hybrids extend the scope of future building blocks towards more versatile qubit structures. Specifically, our focus is on a magnetic system comprising an iron phthalocyanine (FePc) molecule and an Fe atom, resulting in a mixed spin(1/2,1) antiferromagnetically coupled Heisenberg system. This chemical complex shows properties attributed to a spin-1/2 system, which we characterize by driving it into coherently controlled ESR. By using both pump-probe measurements and coherent control techniques, we find a remarkable enhancement in the relaxation time of the hybrid, reaching approximately 1us. This significantly surpasses the values of pristine FePc [1,2] and we attribute this enhancement to the correlation between the two spins in the dimer as well as to enhanced potential scattering. [1] Zhang, X., et al., Nat. Chem. 14 ,59-65 (2022); [2] Willke, P., et al., ACS nano 15, 17859-17965 (2021)

O 70.14 Wed 18:00 Poster C

Spin and charge control of topological end states in chiral graphene nanoribbons — •Leonard Edens[1], Francisco Romero[1], Manuel Vilas-Varela[2], Giuliana Beretta[3], Sofia Sanz[4], Thomas Frederikssen[4], Diego Peña[2], and Nacho Pascual[1] — [1]CIC nanoGUNE, Spain — [2]CiQUS and Universidade de Santiago de Compostela, Spain — [3]Politecnico di Torino, Italy — [4]DIPC, Spain

Chiral graphene nanoribbons can exhibit radical end states due to nontrivial electronic topology. A localized spin fingerprint however remains unobserved as electron transfer to or from the underlying catalyst metal due to work function mismatch causes deviation from half filling, quenching spin. Gold alloyed with rare-earth metals still catalyses on-surface synthesis while exhibiting a lower work function. We show here that (3,2,8)-chiral nanoribbons can be synthesized with atomic precision on the ferromagnetic $GdAu_2$ intermetallic surface, and do not undergo significant charge transfer. By tunneling spectroscopy, we furthermore observe that the occupancy of the two topological end states can be reversibly switched from singly to doubly occupied by lateral manipulation of the nanoribbon across the substrate's moiré lattice. In the doublet case, low-energy features are interpreted as Kondo-screened spin-flip excitations of the π-radical state acted upon by the Gd 4f-mediated exchange bias.

O 70.15 Wed 18:00 Poster C

Paramagnetic resonance of individual organic molecules driven by quantum spin torque — Stepan Kovarik[1], Richard Schlitz[1,2], Aishwarya Vishwakarma[1], •Dominic Ruckert[1], Pietro Gambardella[1], and Sebastian Stepanow[1] — [1]ETH Zürich, Zurich, Switzerland — [2]Department of Physics, University of Konstanz, Germany

It is well-established that time-dependent electromagnetic fields can excite the spin states of a quantum system. In electron paramagnetic resonance (EPR), an oscillating magnetic field B_1 with RF frequency allows coherent control over the spin state of single atoms or molecules.

However, our research shows an alternative mechanism for initiating spin resonance on a single pentacene molecule locally probed in STM.

Here, the mechanism for driving spin resonance is mediated by spin-polarized transport through a locally probed single molecule on the surface. A spin-polarized current oscillating with RF frequency can drive the precession of the localized magnetic moment as theoretically predicted by Shakirov et al. [1]. This process provides a distinct and complementary perspective to the B_1-driven mechanism [2].

Our findings allow the current induced spin initialization of a single spin and thereby hold promise for advancements in molecular-scale electronics and spintronics

O 70.16 Wed 18:00 Poster C

Aging in the self-induced spin glass Nd(0001) — Lorena Niggli[1], •Julian H. Strik[1], Anders Bergman[2], Mikhail I. Katsnelson[1], Daniel Wegner[1], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands — [2]Department of Physics and Astronomy, Uppsala University, Uppsala, Sweden

Elemental neodymium has been shown to be a self-induced spin glass, where glassy behaviour stems solely from the frustrated nature of the magnetic interactions [1]. This contrasts with traditional spin glasses, where the presence of disorder is essential toward realizing glassy behaviour. The magnetic state of Nd(0001) is characterized by a lack of long range order, but exhibits local non-collinear order (Q-states). Upon increasing the temperature, neodymium displays an unusual magnetic phase transition from a self-induced spin glass to a long-range ordered multi-Q phase [2]. Here, we explore the aging behaviour of Nd(0001) in its self-induced spin glass state using spin-polarized scanning tunneling microscopy in varying magnetic fields and variable temperature. We explore how the favourability of the Q-states evolves as we age the system and relate these changes to the preferred structure of the ordered phase. These observations indicate that neodymium may be a multi-well system, which deviates from the traditional energy landscape expected of spin glass systems, thus providing a new platform to study aging dynamics as well as dynamic heterogeneity.

[1] U. Kamber et al., Science 368 (2020).

[2] B. Verlhac et al., Nat. Phys. 18 (2022).